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TRANSPARENT MIXED EPOXY RESIN COMPOSITION AND SAFETY LAMINATED ARTICLES MADE THEREFROM

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The invention relates to an improved epoxy resin composition and to laminar structures employing the composition as the laminant, as well as to the method of making the laminar structure.

This application is a continuation-in-part of co-pending application for U.S. Letters Patent having Ser. No. 783,189, filed Dec. 29, 1958, now abandoned.

The term "epoxy resin," as used herein, refers to a resinous composition employing an epoxylated or epoxidized polyhydric organic compound in the preparation thereof. An epoxylated or epoxidized compound contains at least one oxirane group per molecule. Epoxy resins include liquid or relatively low-melting thermoplastic resins composed of such compounds having an average of more than 1 oxirane group and usually at least an average of 1.5 oxirane groups per molecule and which may be (and are) admixed with a hardening agent while remaining in an uncured state, as well as the solid, thermosetting product obtained by curing the resin/hardening agent admixture. The term "laminar structure," as used herein, refers to at least two flat or contoured sheets or plates integrally bonded in relatively closely spaced, substantially (but not necessarily) parallel, opposing relationship using a solid adhesive composition for the lamination between the sheets or plates. The adhesive composition generally is fluid when applied or injected and is subsequently cured, hardened, or set in situ to a non-fluid state. The term "laminant," as used herein, refers both to the fluid adhesive compositions employed in making laminar structures, and to the solid adhesive composition in the cured state.

A large number of types of laminar articles and structures are widely used. They usually consist essentially of tiers or plies of sheets, panels, plates, and the like of solid wood, glass, plastic, metal, or of pressed, bonded, or matted cellulosic, fibrous, granular, or fragmented material of various types laminated by means of various glues, cements, and the like.

It would be advantageous to provide improved and even more suitable laminants for use in making various types of laminar articles and structures adapted to give satisfactory service for prolonged periods of use under widely differing conditions.

There is a continuous and growing need for improved laminants for specific uses. A particular need exists for a laminant which, in the uncured state, is fluid and flows readily at moderate heat conditions and which cures rapidly and firmly to a thermoset solid. Another particular need exists for a laminant which, when cured, is sufficiently flexible and elastomeric, yet sufficiently strong and tough, to withstand shock and strains induced by impact and abrupt pressure changes, and which possesses stability against deleterious effects of high frequency radiations. A more particular need exists for a laminant, which, when cured, is also of such adhesive nature that it adheres to solid surfaces of materials of the types employed as lamina that the bonds formed between the laminant and the solid surface will not rupture when subjected to strain or shock but will tenaciously adhere even though the lamina itself be broken, thereby rendering the laminar structure shatterproof. A still more par-

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ticular need exists for such laminant which, when cured, is also stable under wide temperature conditions so that it can be subjected both to frigid and tropical temperatures without any detectable lessening of adhesion or cohesion and without any permanent deterioration of desirable properties. A further need exists for a laminant which is also transparent and non-distorting of images and is acceptable for use in laminating transparent articles intended for use in transparent panels and viewing screens.

The present invention meets these needs by providing a resin composition which has low viscosity and which, prior to admixture therewith of a hardening agent, is stable under normal handling conditions including storage, shipping, and the like without detrimental effect. When the resin of the invention is admixed with a curing agent and allowed to stand, it cures readily. This is generally accompanied by substantially no shrinkage nor creation of voids, haze, or objectionable color. It forms an elastomeric, flexible, strong and tough product that is capable of withstanding shock, ray emanations, e.g., those produced by a Crooke's or cathode ray tube, sudden and pronounced pressure changes, physical impact, and extremes of heat and cold. It is transparent and non-distorting. When it is laminated to surfaces of glass, plastic, and the like, it adheres thereto with such tenacity that the bond is retained and unbroken even when the laminar structure is subjected to impact, shock, or sudden pressure change (e.g., an implosion) such as to cause breakage of laminae into innumerable small pieces. Thus, the resin imparts shatterproof properties to the laminar structure.

After the resin/hardening agent admixture has been fully cured, its physical properties remain substantially unchanged for very long periods of time that, often times, extend far beyond the practical need therefor.

The resin is well suited for casting, plying, and layering, particularly where an adhesive of superior properties is desired. It is also useful for coating, impregnating, and the like. It is, as stated, especially suitable for laminating transparent sheets, e.g., glass or plastic.

Glass laminar structures employing layers (up to 0.1 inch or so thick) of the resin of the invention as the laminant exhibit all the above mentioned desirable properties. For example, typical laminar structures employing the resin of the invention in a thickness of about 0.06 inch, a thickness especially suitable for laminating, show a bond strength to the glass of over 135 pounds per square inch (p.s.i.). Such typical specimens of the glass laminar structure of the invention can be repeatedly and alternately subjected to temperatures of -50°F. and 180°F. and a relative humidity of 95 percent at 100°F. for many months with no accompanying or resulting noticeable deteriorating effects. Thus, such exposure occasions no "pock" marks, "worm" tracks, or other indications of weakening of adhesive or cohesive bonds (or in some instances, breaking of the lamina itself). Such defects are common for most resins when employed as a laminant due, among other things, to lack of adequate cohesive or adhesive strength and of satisfactory flexibility. The strain inducing the above undesirable effects is understandable when it is borne in mind that the coefficient of thermal expansion of glass is about 0.9×10^{-4} whereas, that of plastics, although varying, is substantially greater, 0.5 to 7×10^{-3} being illustrative thereof.

Standard A.S.T.M. tests designed to ascertain the percent elastic recovery and percent elongation at break are indicative of flexibility. Typical specimens of the invention, when so tested, have elastic recoveries as high as 95 percent and elongations at break, at 77°F. , of over 200 percent. It is understood that the resin of the invention may have lower values than those above, e.g., of